Spin fluctuations in the quasi-two dimensional Heisenberg ferromagnet GdI_2 studied by Electron Spin Resonance

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The spin dynamics of GdI_2 have been investigated by ESR spectroscopy. The temperature dependences of the resonance field and ESR intensity are well described by the model for the spin susceptibility proposed by Eremin *et al.* [Phys. Rev. B 64, 064425 (2001)]. The temperature dependence of the resonance linewidth shows a maximum similar to the electrical resistance and is discussed in terms of scattering processes between conduction electrons and localized spins.

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I. INTRODUCTION

Recently, the phenomenon of giant magnetoresistance has attracted considerable interest, not only in the manganites[1] but also in spinel-type[2] or intermetallic compounds.[3,4] Gadolinium diiodide came into the focus of solid-state research because of reports of giant negative magnetoresistance effects at the ferromagnetic phase-transition close to room temperature.[5,6] GdI_2, which contains formally divalent Gd with an electronic configuration $4f^7 5d^1$, is a correlated narrow d-band metal revealing ferromagnetism. GdI_2 has first been synthesized by Mee and Corbett.[7] It crystallizes in the close-packed hexagonal 2H-MoS_2 structure (space group $P6_3/mmc$), which has a strongly two-dimensional (2D) character: the rare-earth layers, in which each Gd atom is surrounded by six nearest Gd neighbours, are separated by two I atom layers. The stacking sequence is built up along the c-axis (see Fig. 1). A first study of the magnetic properties was performed by Kasten *et al.* characterizing GdI_2 as a ferromagnet with a Curie temperature near room temperature,[8] at which the Gd moments are aligned perpendicular to the c-axis.

Using a semi-phenomenological approach, the large magnetoresistance has been explained in terms of a d-f exchange model: The anomalous peak of the resistance and the negative magnetoresistance are due to a strong scattering of the d-derived conduction electrons by the localized $4f$ electrons.[9] Furthermore, also from its magnetic properties GdI_2 is highly interesting. Gd exhibits a half-filled $4f$-shell with a spin-only moment of 7 μ_B. Any spin-orbit coupling is expected to be weak and in the paramagnetic phase GdI_2 should be a good realization of a 2D-Heisenberg ferromagnet. Theoretically, a 2D Heisenberg ferromagnet is expected to order at 0 K only. Of course, interplane interactions cannot be neglected and GdI_2 reveals bulk ferromagnetic order below 290 K with an ordered moment of 7.33 μ_B. The excess moment of 0.33 μ_B, when compared to the spin-only moment of the Gd $4f$ shell, obviously results from a strong polarization of the $5d$ conduction band.[6]

II. SAMPLE PREPARATION AND EXPERIMENTAL DETAILS

GdI_2 has been prepared in a solid-state reaction of GdI_3 and Gd metal powder at 1100 K for three weeks in a sealed Ta tube jacketed with an evacuated silica ampoule. Details of the sample preparation are given in references 5 and 6. The structure has been determined by x-ray powder diffraction and lattice parameters $a =$

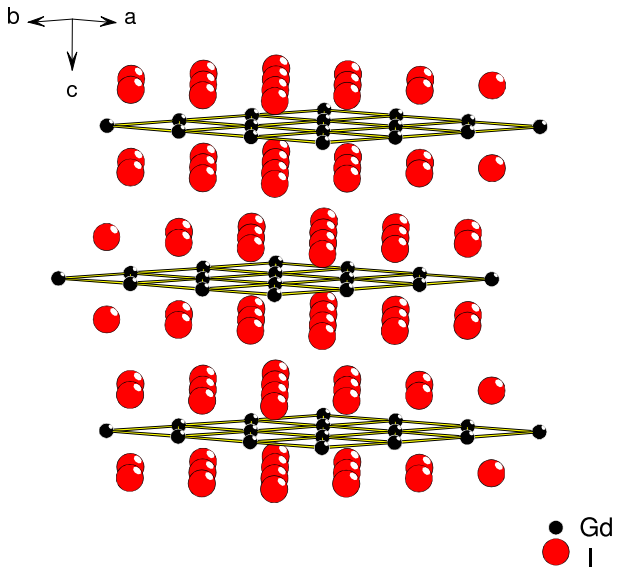

FIG. 1: Crystal structure of GdI_2 (space group $P6_3/mmc$).

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0.40775(4) nm and $c = 1.5041(1)$ nm were obtained. For the samples under investigation, from an analysis using modified Arrotts plots the ferromagnetic phase-transition temperature has been determined as $T_C = 276$ K.[5]

ESR measurements were performed in a Bruker ELEXSYS E500 CW-spectrometer at X-band frequencies ($\nu \approx 9.47$ GHz) equipped with a continuous N_2-gas-flow cryostat in the temperature region $80 < T < 600$ K. Note that the samples are very sensitive to exposure to air and humidity. Therefore the whole procedure of powdering the polycrystalline samples and placing them into quartz tubes had to be undertaken in He atmosphere. Then the tubes were sealed in He atmosphere, too. ESR detects the power P absorbed by the sample from the transverse magnetic microwave field as a function of the static magnetic field H. The signal-to-noise ratio of the spectra is improved by recording the derivative dP/dH using lock-in technique with field modulation.

III. EXPERIMENTAL RESULTS AND DISCUSSION

A. ESR spectra

ESR spectra obtained for GdI_2 in the paramagnetic regime at different temperatures are displayed in Fig. 2. The spectra consist of a broad, exchange-narrowed resonance line, which is well fitted by a single Dysonian line shape.[10] Although an anisotropy of the resonance line cannot be excluded due to the two-dimensional character of the system, no such indications can be deduced from the powder spectra in GdI_2. In general, powder spectra exhibit a characteristic pattern due to the random distribution of the orientation of the grains, if resonance field and linewidth are anisotropic.[11] The fact that the spectra are well described by a single Dysonian line indicates that an anisotropy, if existing, is smaller than the observed linewidth. As in the present compound the linewidth ΔH is of the same order of magnitude as the resonance field $H_{\rm res}$, both circular components of the exciting linearly polarized microwave field have to be taken into account. Therefore the resonance at the reversed magnetic field $-H_{\rm res}$ has to be included into the fit formula for the ESR signal given by

$$\frac{dP}{dH} \propto \frac{d}{dH} \times \left\{ \frac{\Delta H + \alpha(H - H_{\rm res})}{(H - H_{\rm res})^2 + \Delta H^2} + \frac{\Delta H + \alpha(H + H_{\rm res})}{(H + H_{\rm res})^2 + \Delta H^2} \right\} \quad (1)$$

This is an asymmetric Lorentzian line, which includes both absorption and dispersion, with α denoting the dispersion-to-absorption ratio. Such asymmetric line shapes are usually observed in semiconductors[12] and metals,[10] where the skin effect drives electric and magnetic microwave components out of phase and therefore leads to an admixture of dispersion into the absorption spectra. For samples small compared to the skin depth one expects a symmetric absorption spectrum ($\alpha = 0$), whereas for samples large compared to the skin depth absorption and dispersion are of equal strength yielding an asymmetric resonance line ($\alpha = 1$). An additional contribution to the asymmetry of the resonance line can arise from the fact that ΔH is of the same order of magnitude as $H_{\rm res}$, because then not only the overlap with the resonance at $-H_{\rm res}$ but also the mutual coupling via the nondiagonal elements of the dynamic susceptibility influences the lineshape.[13] As can be seen in Fig. 2 the asymmetry of the resonance remains almost constant for higher temperatures, while on approaching magnetic order from above the resonance strongly shifts to lower fields and concomitantly the asymmetry increases significantly due to the increasing overlap with the corresponding resonance at $-H_{\rm res}$. The temperature dependence of the parameter α is shown in the inset of Fig. 3(b).

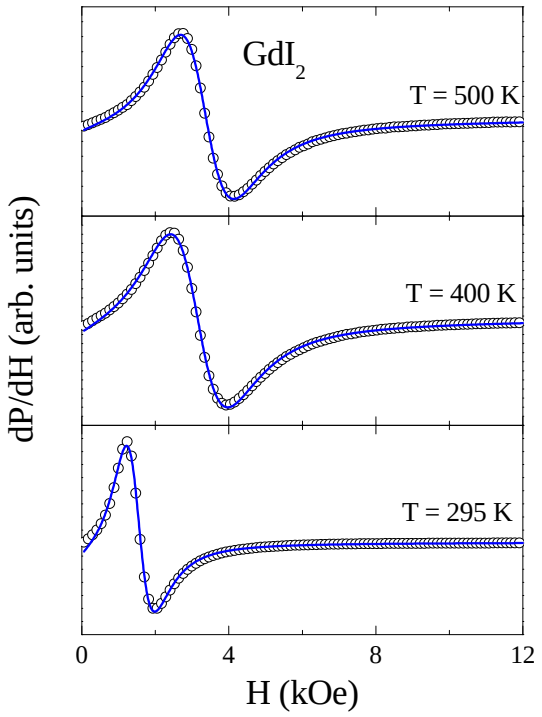

FIG. 2: ESR spectra of GdI_2 for different temperatures illustrating the line shift and linewidth changes. Solid lines are fits obtained by using a Dysonian lineshape according to Eq. (1).

Figure 3 shows the temperature dependences of the main spectral ESR parameters of GdI_2, namely (a) the intensity $I_{\rm ESR}$, (b) the resonance field $H_{\rm res}$, and (c) the half-width-half-maximum linewidth ΔH, which will be discussed in the following.

B. ESR intensity and resonance field

In principle, $I_{\rm ESR}$ measures the spin susceptibility of the Gd spins and is approximated by $I_{\rm ESR} \propto A\Delta H^2$,

where A denotes the amplitude of the field-derivative of the absorption spectrum. In metals, however, one has to account for the finite skin depth δ of the microwave into the sample, which depends on the resistivity ρ and the ESR frequency ω as $\delta = (\rho/\mu_0\omega)^{0.5}$. As for GdI_2 only the resistance R but not the specific resistivity ρ is available at present, the absolute skin depth cannot be calculated. Note that the resistance in the paramagnetic regime exhibits a broad maximum,[6] and, therefore, the changes in the skin depth are negligible, so that one probes always the same fraction of the sample. As we are only interested in relative changes of the spin susceptibility, the use of the above approximation for $I_{\rm ESR}$ is justified. $I_{\rm ESR}$ decreases monotonously with increasing temperature. Following the analysis of spin fluctuations of localized $4f^7$ electrons by Eremin *et al.*[9] for $T > T_{\rm C}$, we use the random-phase-approxomation-like approach to the dynamic spin susceptibility (depending on the wavevector $\mathbf{q}$ and the frequency ω) given by

$$\chi_{\rm spin}(\mathbf{q},\omega) = \frac{\chi_{\rm 2D}(\mathbf{q})}{1 - J_\perp(\mathbf{q})\chi_{\rm 2D}(\mathbf{q})} \frac{i\gamma(\mathbf{q},\omega)}{\omega + i\gamma(\mathbf{q},\omega)}, \quad (2)$$

where $\gamma(\mathbf{q},\omega)$ constitutes the damping function of the spin fluctuations, and $J_\perp(\mathbf{q})$ the ferromagnetic inter-layer exchange constant. Evaluation of the imaginary part of Eq.(2) at $\mathbf{q} = 0$ and using the temperature dependence of the 2D spin susceptibility $\chi_{\rm 2D}$ given by

$$\chi_{\rm 2D}(T) = \frac{1}{6\sqrt{3}SJ_\parallel} \exp\left[\frac{16\sqrt{3}S^2J_\parallel}{T}\right], \quad (3)$$

results in

$$\chi_{\rm spin}(T) \propto \left(\exp\left[-\frac{16\sqrt{3}S^2J_\parallel}{T}\right] - \frac{J_\perp}{6\sqrt{3}SJ_\parallel}\right)^{-1}, \quad (4)$$

with the Gd spin $S = 7/2$ and the ferromagnetic exchange constant $J_\parallel$ within the Gd layers. Note that the determination of absolute values of the susceptibility using Eq. (2) cannot easily be undertaken, as the 'form of the function $\gamma(\mathbf{q},\omega)$ for finite ω is unknown'.[9]

The best fit obtained using Eq. (4) is displayed as a solid line in Fig. 3(a). Within experimental uncertainties, the data can be well described by this approach with fit parameters $J_\parallel = 4.9(1)$ K and $J_\perp/J_\parallel = 0.035(3)$. These values are in good agreement with the estimates $J_\parallel \approx 6$ K and $J_\perp/J_\parallel \approx 0.03$ obtained from the analysis of the Curie temperatures by Eremin and coworkers.[9] Deviations from the fitting curve are largest in the temperature range $300 < T < 350$ K, where the linewidth is reduced by a factor of three. The approximation $I_{\rm ESR} \propto A\Delta H^2$ strictly holds only for $\Delta H \ll H_{\rm res}$ but can also be used for larger ΔH, if ΔH changes only slightly as in our case above 350 K. Therefore, the largest uncertainty in $I_{\rm ESR}$ certainly occurs where the changes in linewidth are largest.

The resonance field $H_{\rm res}$ or the effective g-value $g_{\rm eff}$=$h\nu/(\mu_{\rm B}H_{\rm res})$ provides information about the local static magnetic field at the Gd site. The asymptotic high-temperature value of the resonance field $H_{\rm res}(\infty) = 3.4$ kOe corresponds to an effective $g_{\rm eff}$-value of $g = 1.99$ similar to the value observed for Gd^{3+} ions in insulators,[14] as expected for vanishing orbital moment ($L = 0$) of the half-filled $4f$ shell. At about 400 K the resonance field starts to decrease dramatically towards the ferromagnetic ordering at $T_{\rm C} = 276$ K.[6] Considering the strong influence of the ferromagnetic fluctuations on the resonance shift on approaching $T_{\rm C}$ from above and the quite large values of the susceptibility $\chi > 0.1$ emu/mol for $T < 400$ K,[9] demagnetization effects cannot be neglected. Due to the relation between intrinsic and observed susceptibility

$$\frac{1}{\chi_{\rm obs}} = \frac{1}{\chi_{\rm int}} + N, \quad (5)$$

where N denotes the demagnetization factor of the order of 4π along the direction of the applied magnetic field, one expects important corrections. Therefore we use the famous formula derived by Kittel[15] for ferromagnetic resonance in an ellipsoidal sample given by

$$\frac{\omega_0}{\gamma} = \sqrt{\left[H_{e0} + (N_x - N_z)\,M_0\right]\left[H_{e0} + (N_y - N_z)\,M_0\right]}, \quad (6)$$

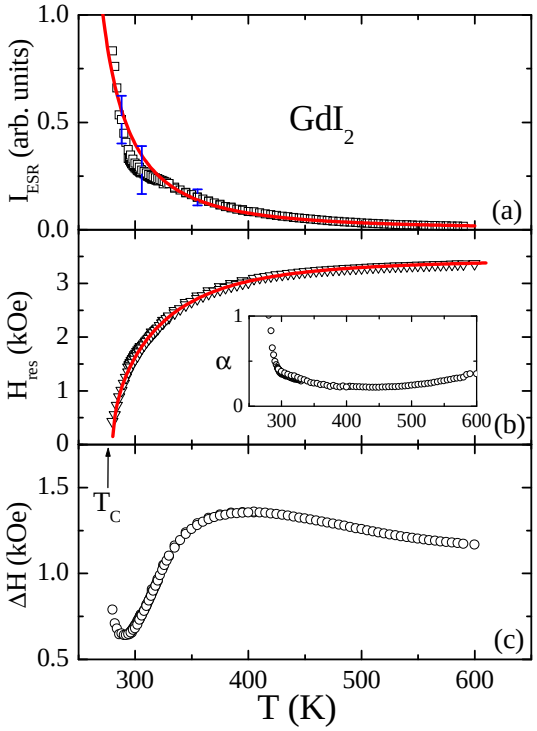

FIG. 3: Temperature dependence of (a) the ESR intensity $I_{\rm ESR}$, (b) the resonance field $H_{\rm res}$ including the dispersion-to-absorption ratio α in the inset, and (c) the ESR linewidth ΔH in GdI_2. Solid lines in (a) and (b) represent fits obtained by using Eqs. (4) and (8) as described in the text, respectively.

where ω_0 denotes the resonance frequency, γ the gyromagnetic ratio, H_{e0} the external magnetic field applied parallel to the z-direction, N_i the demagnetization factors, and M_0 the magnetization of the sample. Approximating the shape of the sample as a cylindrical disk with finite thickness ($N_y = N_z \ll N_x, \quad N_x - N_z =: N_{\rm res}$) and with the external magnetic field H_{e0} applied within the disk's plane one derives

$$H_{e0} = H_0/\sqrt{1 + N_{\rm res}\chi} \tag{7}$$

by using $M_0 = \chi H_{e0}$ and $H_0 = \omega_0/\gamma$. Applying the proportionality for $\chi_{\rm spin}(T)$ given by Eq. (4) the following fit formula for the resonance field $H_{\rm res} = H_{e0}$ emerges:

$$H_{\rm res}(T) = \frac{H_0}{\sqrt{1 + A\left(\exp\left[-\frac{16\sqrt{3}S^2 J_\parallel}{T}\right] - \frac{J_\perp}{6\sqrt{3}SJ_\parallel}\right)^{-1}}} \tag{8}$$

The dimensionless parameter A includes the demagnetization factor and the unknown prefactors of the susceptibility (see above). The resulting fit curve (solid line in Fig. 3(b)) yields parameters $A = 2.2(1) \times 10^{-3}$, $H_0 = 3468(3)$ Oe, $J_\parallel = 5.6(3)$ K, and $J_\perp/J_\parallel = 0.039(2)$. This good agreement with the ESR intensity and the reported values by Eremin and coworkers[9] corroborates the consistency of our approach.

C. Linewidth

On decreasing temperatures the resonance broadens and the linewidth exhibits a broad maximum at about 390 K (see Fig. 3(c)). Towards lower temperatures the linewidth decreases and shows a minimum before it abruptly increases again due to inhomogeneous broadening above the ferromagnetic transition. Towards the highest temperatures the linewidth seems to reach a constant value at about 1.1 kOe. Already at first sight one recognizes that $\Delta H(T)$ correlates with the temperature dependence of the electrical resistance, both of which are shown in Fig. 4. It has been proposed that the maximum in $R(T)$ results from strong scattering of the $5d$-derived conduction electrons at the localized $4f^7$-electrons due to a peculiar topology of the Fermi surface in this compound.[5] The conduction electrons in GdI_2 occupy a narrow $5d_{z^2}$ band and the carriers at the Fermi level are close to a Γ point of the first Brillouin zone. Hence, critical ferromagnetic fluctuations influence the magnetic part of the resistivity very strongly.

The ESR linewidth $\Delta H(T)$ generally corresponds to the transverse spin relaxation rate $1/T_2$ due to local fluctuating fields, which in insulators originate from anisotropic interactions like dipole-dipole interaction between the localized spins, hyperfine fields from the nuclei, and the crystal electric field of the surrounding ligands. In good metals, which contain both localized magnetic moments and delocalized conduction electrons, the interaction between them provides an additional important

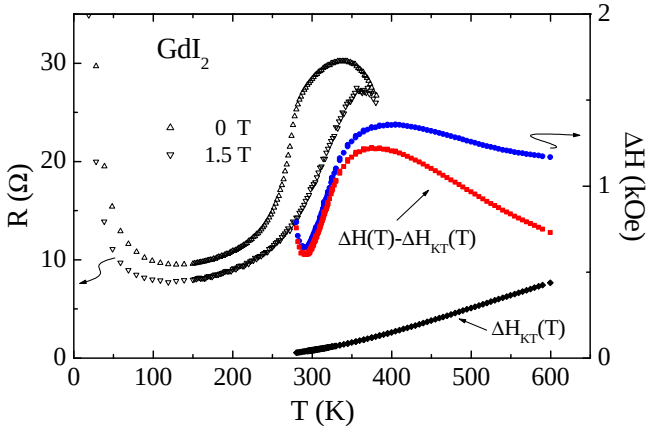

FIG. 4: Temperature dependence of the ESR linewidth together with the temperature dependence of the resistance for GdI_2 in zero-magnetic field and in an applied field of 1.5 T taken from Ref. 5. The contribution $\Delta H_{\rm KT}$ has been estimated as described in the text and subtracted from the linewidth data to highlight the contribution of the d-f scattering.

relaxation channel that usually dominates the broadening mechanism. Regarding the resistance values, GdI_2 cannot be looked upon as a good metal and, hence, we will discuss both line-broadening contributions in the following:

In exchange coupled systems the isotropic Heisenberg exchange J (here $J_\parallel \approx 6$ K) leads to an exchange narrowing of the ESR spectrum into a single line, which is usually described by the Kubo-Tomita approach,[16]

$$\Delta H_{\rm KT} \simeq \frac{M_2}{H_{\rm ex}} \tag{9}$$

with the second moment M_2 of the resonance line and exchange field $H_{\rm ex} \propto J$. The temperature dependence of the linewidth in this case is given in the high-temperature approximation by

$$\Delta H_{\rm KT}(T) = \frac{\chi_0(T)}{\chi(T)}\Delta H_\infty \tag{10}$$

with the free Curie susceptibility $\chi_0 = C/T$, where $C = Ng^2\mu_{\rm B}^2 S(S+1)/3k_{\rm B}$ denotes the Curie constant of the Gd ions with $S = 7/2$, and the measured dc-susceptibility $\chi(T)$.[17] As $\chi_0(T)/\chi(T) \to 1$ for $T \to \infty$, the temperature independent parameter ΔH_∞ can be identified with the high-temperature limit of the ESR linewidth. To estimate this contribution to the ESR linewidth, we follow the arguments by Sperlich *et al.* for the case of GdB_6.[18] The authors conclude that the main contribution to the resonance linewidth is given by the dipole-dipole interaction of the rare-earth spins contributing about 0.3 kOe to the linewidth for GdB_6, which has an exchange constant of about 1 K. With $J \approx 6$ K in GdI_2 this value is about 50 Oe and too small to fully account for the observed high-temperature value of the linewidth. However, it

cannot be excluded that the strong two-dimensionality of the system effectively reduces the exchange-narrowing effect resulting in a larger contribution of the dipole-dipole interaction.[19]

Due to very short correlation times $\tau \ll \omega_0$ in metals, longitudinal T_1 and transversal spin relaxation time T_2 are equal.[20] Hence, in this case the ESR linewidth directly measures the spin-lattice relaxation rate $1/T_1$. In the case of a half-filled $4f$-shell with spin $J = S = 7/2$ the orbital momentum of Gd^{3+} vanishes and a direct relaxation to the lattice is not possible. Nevertheless, the energy is transferred to the lattice by scattering of the conduction electrons at the localized moment of the $4f$ shell. This is the well-known Korringa relaxation which gives rise to a linear increase of the Gd linewidth with increasing temperature.[21] This behaviour is typically observed for Gd ions diluted in usual metals and also in many concentrated Gd-based transition-metal compounds above magnetic order.[22,23] The slope of the linear increase is determined by the electronic density of states at the Fermi level $\propto N^2(E_F)$. Deviations from this canonical behaviour have already been found from strongly correlated electron systems like heavy-fermion compounds, where the density of states is strongly modified near E_F.[24] In these compounds, both the resistivity and Gd-ESR linewidth show a typical non-linear temperature dependence with a maximum near the characteristic temperature $T^* \ll E_F/k_B$ for the screening of the Kondo ions by the conduction electrons.

In GdI_2 we do not have any Kondo effect with its antiferromagnetic screening of the localized moments,[25] but instead there are strong ferromagnetic fluctuations due to the exchange interaction between the Gd $5d$ band and the localized $4f$ shell. Like in the case of the Kondo effect the electrical resistance is dominated by these fluctuations and vice versa the spin relaxation of the Gd $4f$ shell is strongly affected. Therefore the maxima of resistance and linewidth which coincide approximately indicate the characteristic temperature of the ferromagnetic fluctuations in the range $T_C < T^* < \Theta_{CW} = 412$ K. The Curie-Weiss temperature Θ_{CW} was obtained from the high-temperature regime of the susceptibility reported in Ref. 9.

To accentuate the contribution due to the ferromagnetic spin fluctuations it seems useful to subtract the contribution of the high-temperature relaxation. Both possible contributions to the high-temperature linewidth, dipole-dipole interaction following Eq. (10) or a linear Korringa increase, if metallic properties dominate, would yield a monotonic increase with increasing temperature. Subtracting such a contribution results, hence, in a more pronounced maximum in agreement with the maximum of the resistance. To illustrate this we assumed a high-temperature value $\Delta H_\infty = 1.1$ kOe corresponding to the linewidth at the highest experimental temperatures and simulated the Kubo-Tomita contribution Eq. (10) using the susceptibility data from Ref. 9.

Compared to $R(T)$, however, the linewidth maximum is shifted to higher temperatures. This can be attributed to the finite external magnetic field in the ESR experiment, which is swept over a range from 0 to 1.5 T. Field-dependent resistance measurements evidence a significant shift of the maximum to higher temperatures with increasing strength of the magnetic field.[6] From theoretical estimates, it follows that the maximum in $R(T)$ shifts by approximately 10 % in external magnetic fields of 1 T,[9] hence accounting for the observed differences between ESR linewidth and resistance.

Interestingly, a correlation between the temperature dependence of $\Delta H(T)$ and $R(T)$ has also been observed in the intermetallic compound Gd_2PdSi_3.[26] Similar to GdI_2 this compound is characterized by a large magnetoresistance effect and, though showing antiferromagnetic ordering at $T_N = 21$ K, exhibits a positive Curie-Weiss temperature $\Theta_{CW} = 25$ K evidencing the existence of strong ferromagnetic correlations.[3] On approaching T_N both resistivity and ESR linewidth attain a minimum at $T \approx 2T_N$, followed by a similar increase of both quantities towards the Néel temperature. It seems likely that this feature could be due to similar scattering processes as in GdI_2, especially, because the occurrence of anisotropic magnetoresistance effects and anomalies in the Hall resistivity strongly suggest the existence of an anisotropic Fermi surface and its reconstruction across the metamagnetic anomaly observed in Gd_2PdSi_3.[27] To establish such a scenario, however, a theoretical analysis of the ferromagnetic correlations in Gd_2PdSi_3 is highly desirable.

IV. CONCLUSIONS

In summary, we find that in GdI_2 the temperature dependence of the ESR linewidth correlates with the resistance and conclude that the linewidth is dominated by scattering processes of conduction electrons at the localized f electrons. Both the resonance field and the ESR intensity can be well described by a phenomenological approach taking into account ferromagnetic spin fluctuations of the f electrons. For the resonance field and the ESR intensity the analysis yields in- and out-of-plane exchange constants $J_\parallel = 5.6; 4.9$ K and $J_\perp/J_\parallel = 0.039; 0.035$, respectively, which are in excellent agreement with previous estimates.

Acknowledgments

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